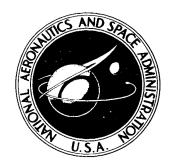
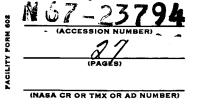
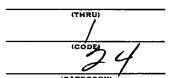
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GAS DENSITY MEASUREMENTS USING LIGHT SCATTERING

by Stuart L. Seaton

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SUMMARY

Experiments are described and results discussed for measuring gas density by means of laser light scattered by gases. Rayleigh theory is confirmed for the experimental conditions encountered. Polarizabilities experimentally obtained agree satisfactorily with those from other sources based on refractive indices of the test gases. It is concluded that the feasibility of the method has been established.

INTRODUCTION

It is often desirable in contemporary hypersonic low-density, short-run-time facilities to make measurements of free-stream gases by probeless means. Because temperatures are high, and response times needed are often very short, it is difficult to permit physical configurations (rakes, probes, and the like) in the stream itself. Often, the presence of probes both changes the flow and alters the state of the gas and thus interferes with the intended measurements. Consequently, means have been sought to devise and apply indirect approaches of a probeless type so that the density of the free stream can be determined without the stream being affected.

One of the attractive ways to measure free-stream density centers around the phenomenon of scattering of light by a gas, as propounded originally by Strutt (later Lord Rayleigh) (ref. 1), but using more modern tools such as the laser and the photomultiplier tube. The laser brings intense monochromatic light of small angular spread to bear on the gas under study. The photomultiplier tube has very great sensitivity and the fast response needed to examine light scattered from a small volume of gas during short time intervals. Less intense sources and less sensitive detectors have heretofore limited the usefulness of the method.

Investigations were, therefore, undertaken to see whether this approach to measurement of gas density might prove feasible and accurate. The light scattered from several gases and gas mixtures over a range of pressures was studied in a static test chamber and the data were examined in terms of scattering theory. The results are described in this report. The research reported herein was carried out by the author and by

Courtney E. Russ, Jr., who was responsible for the laboratory aspects of the investigation and for reduction of the raw data.

SYMBOLS

A_{O}	attenuator factor, incident light
A_S	attenuator factor, scattered light
a_{O}	output voltage, incident light, volts
a_{S}	output voltage, scattered light, volts
I _o	incident light intensity, watts/meter ²
I_S	scattered light intensity, watts/meter ²
m	refractive index
N	number density, cm ⁻³
r	distance from scatterer to receiver, cm
α	polarizability, cm ³
θ	angle between incident and scattered light
λ	wavelength, cm
ν	frequency, hertz

LABORATORY APPARATUS

Test Chamber

A test chamber was constructed for the purpose of conducting a controlled experiment aimed at determining gas density, accuracy of measurement, repeatability, and other factors essential for the design of a gas-density measuring tool for application to hypersonic free streams. A number of gases were introduced into the test chamber and each was studied separately.

Figure 1 is a sketch of the apparatus giving principal dimensions and angles. Figure 2 shows the experimental equipment and arrangements. Essentially, the laboratory model consisted of a chamber of about $10^5~{\rm cm}^3$ capacity lined with black velvet and containing baffles to suppress internally scattered light. The laser beam entered the chamber through a glass vacuum barrier normal to the beam direction. The glass barrier was originally placed at the chamber inner surface but this arrangement produced so much scattered light in the chamber that the glass was moved back 21 cm and baffles placed between it and the chamber wall to eliminate most of the unwanted light. After traversing the chamber, the laser beam passed a baffle and went out through an exit port of glass set at an angle of 20^0 to the beam normal. Thence, the beam was dissipated in a long black baffle box.

The glass plate just in front of the laser served, as noted, to isolate the gas in the test chamber. This plate is also part of the monitoring system since the edge of the plate is viewed by a photomultiplier tube through neutral density filters which attenuate the light scattered by the glass plate to a level approximating intensity of light scattered by the gas in the test chamber.

As can be seen in figure 1, the receiver beam intercepts the laser beam about 33 cm into the chamber and develops an angle of 22° with respect to the line of the laser beam (158° with respect to the forward direction of the laser beam). Thus, the received energy is largely backward scattered. The receiver looks into a black baffle box.

Both receiver and monitor tubes viewed their sources through stacked filters Wratten Nos. 29 and 35 which constitute low pass filters with the cutoff just to the high-frequency side of the laser frequency ($\lambda = 6943$ Å, and $\nu = 4.34 \times 10^{14}$ hertz). Use of these filters reduced the unwanted light by about a factor of two. Narrow pass interference filters would have been an improvement for further reducing the unwanted light.

The common volume of intersection of receiver and transmitter beams was approximately $0.25~{\rm cm}^3$.

Apparatus Designations

The laser used was a Raytheon LH-2-C ruby rod device rated at three joules, not Q-switched, and pumped at about 800 joules. Liquid nitrogen cooling was employed to keep the ruby from overheating. Dry nitrogen at room temperature was circulated across the surfaces of the monitor glass and the exit face of the ruby to prevent frosting of these surfaces from ambient moisture. The laser pulse length was of the order of 800 microseconds.

Monitoring was done with an EMI 9558-A photomultiplier tube viewing the edge of the vacuum isolating glass normal to the laser beam. In other words, light scattered in

the glass and issuing through the \underline{edge} of the glass provided the light used to monitor the laser output.

Light scattered by the test gas was viewed by an RCA 7265 photomultiplier tube. A rack and slide device was provided on this receiver so that the axis of the receiver "beam" could be caused to coincide exactly with the laser beam; thus, the common volume of intersection was developed.

Outputs from the photomultipliers were applied to a calibrated dual-beam oscilloscope through its amplifiers, and the beam traces were photographed. Figure 3 is a typical record of the oscilloscope traces.

Controls

Classically, the amount of light scattered by a gas is linearly proportional to the density of the gas. Since it was not feasible to make absolute measurements, a technique was adopted wherein the ratio of scattered to incident intensity was determined.

The gas pressure in the test chamber was measured with an absolute pressure gage. A Hastings gage was used below about 20 millitorr. In addition, a record was kept of ambient humidity, temperature, and barometric pressure. For some reason not understood, the RCA 7265 photomultiplier tube system (scattered light) was sensitive to local barometric pressure by an amount of 0.026 volt per cm Hg. Otherwise, the systems were not affected by ambient conditions, nor should they have been since the test chamber was of rather rugged construction.

Experiments were made with dry air, carbon dioxide, nitrogen, helium, oxygen, and argon. The pressures ranged between about 50 millitorr at the lowest and about 650 torr maximum.

EXPERIMENTAL RESULTS

Experimental Data

Figure 4(a) gives results of measurements obtained by using filtered, dry, natural air. The ordinate is the ratio of phototube outputs and the abscissa, the chamber pressure.

Figures 4(b) to 4(f) show results for the following gases, respectively: carbon dioxide, argon, nitrogen, helium, and oxygen. Each curve was developed by drawing a mean line through the cluster of 10 points at each of the following pressures: 100 millitorr, 200 torr, 400 torr, and 600 torr. Extrapolation to 1 atmosphere is also shown. On each graph the scatter about the mean is given by vertical bars.

Since it was desired to measure the gas pressure, it was necessary to examine the accuracy with which the pressure could be deduced from measurement of intensity of light scattered by the gases.

Each gas has a characteristic curve slope of the variation of pressure with scattered light intensity. The slope of this curve is proportional to the polarizability of the particular gas. As can be seen in figure 5, the scatter in observed light intensity, when projected on the curve gives a corresponding range in pressure. For a given scatter in light intensity, the pressure uncertainty is larger for gases with small curve slope (small value of polarizability) than for gases with greater slope (larger values of polarizability).

The scatter in the ratio of phototube outputs was everywhere about the same. Thus, the accuracy with which pressure could be deduced was mainly a function of the pressure and of the polarizability. (See fig. 6.) The best data are those of the lower curve and the least accurate measurements, the upper bound. Helium, which has such a small curve slope as to render any individual measurement meaningless is not included, although mean values appear to be very good as will be seen subsequently.

Scaling Techniques

Two methods of scaling the records were tried. Unless otherwise noted, the peak values were used and the ratio of these readings accepted as the data. However, the area under the curves was also measured for comparison. The latter method was more accurate, but was too time consuming for routine use.

Human scaling error for peak values, as determined by comparing results of four independent scalings among three persons, was found to be about 5 percent.

DISCUSSION OF EXPERIMENTAL RESULTS

Theory

Van de Hulst (refs. 2 and 3) notes that the Lorentz-Lorenz formula relating refractive index to polarizability:

$$4\pi\alpha N = \frac{3(m^2 - 1)}{m^2 + 2} \tag{1}$$

is based on the following conditions:

- (a) Many particles exist in a sphere of radius λ
- (b) Mutual distances of the particles are small compared with λ
- (c) Particle size is much smaller than λ

- (d) Particles are isotropic scatterers
- (e) The field to which each particle is subjected is due to the scattered fields of many other particles at all sides (fields essentially in phase with incident field)
- (f) The statistical effect of these fields gives rise to the Lorentz-Lorenz formula. However, when the mutual distances are large compared with λ , when the particles are small with respect to λ , and when m approaches unity, the following expression is applicable:

$$m - 1 = 2\pi\alpha N \tag{2}$$

For ruby laser light (6943 Å) and for pressures of less than 1 atmosphere, the following numbers are appropriate:

Wavelength, cm	$\approx 7 \times 10^{-5}$
Interparticle distance, cm	
Particle diameter, cm	$\approx 1 \times 10^{-8}$
Index of refraction	<1.00050

It appears therefore that expression (2) is more nearly applicable to these experiments than is expression (1) over most of the range. However, at higher pressures (greater than 1 atmosphere), the index of refraction increases and the interparticle distance diminishes. These changes tend to approach the domain of validity of expression (1). It may be, then, that near 1 atmosphere, a changeover begins to appear, and if experiments were performed at pressures much greater than 1 atmosphere, a clearly defined alteration in the slope of the curve would become evident. As it is, the expected difference between values computed from expressions (1) and (2) at 1 atmosphere is around 1 percent and is too small to be distinguished in these experiments.

From the Rayleigh expression (refs. 2 and 3) for light scattered per unit volume,

$$\frac{I_S}{I_O} = \frac{8(1 + \cos^2\theta)\pi^4\alpha^2N}{r^2\lambda^4}$$
 (3)

and for θ , r, and λ constant, as they were in these experiments, it would be expected that I_s/I_0 would vary in proportion to α^2N . If α for each gas is constant under these experimental conditions, the relationship between a_s/a_0 and N should be linear; it was found to be linear within the experimental error of observation, as can be seen from figure 4.

Equivalent System

The laboratory system can be viewed schematically as in figure 7. The attenuators A_O and A_S have negative as well as positive gains. They are composed of an optical part ahead of the phototubes having negative gain, and an electrical part with positive gain following the phototubes. These parts are combined in the figure for purposes of simplicity, since the net effect is to vary the total gain in each channel as desired. The notations a_O and a_S refer to the amplitudes of the "meter" readings at the output ends of the system; then,

$$\begin{vmatrix}
A_O I_O = a_O \\
A_S I_S = a_S
\end{vmatrix}$$
(4)

Initially, the system was adjusted by changing A_O and A_S so that

$$\mathbf{a_0} = \mathbf{a_S} \tag{5}$$

for a datum value of $\,N\,$ in the scattering volume. The value of $\,N\,$ was then changed and the "meter readings" of $\,a_{O}\,$ and $\,a_{S}\,$ determined. It is to be noted that $\,I_{O}\,$ is nearly constant and thus $\,A_{O}\,$ is left unchanged unless $\,I_{O}\,$ is sensibly altered for some reason or other. The value of $\,A_{S}\,$ is changed as $\,N\,$ is changed in order to maintain the condition $\,a_{O}\,\approx a_{S}.\,$

Instrument Constant

Obviously, $I_O >> I_S$ and the instrument constant for air at a reference pressure of 1 atmosphere at standard temperature will now be evaluated.

If equation (3) is recalled and it is noted that the term in brackets is constant for this purpose,

$$\frac{I_{S}}{I_{O}} = \alpha^{2} \left[\frac{8\pi^{4}(1 + \cos^{2}\theta)N}{r^{2}\lambda^{4}} \right]$$
 (6)

Then for $\alpha = 1.79 \times 10^{-24}$ computed from equation (2) by using m - 1 = 2.93×10^{-4} from tables of refractive index (refs. 4 and 5) of air at optical wavelengths (compare ref. 6),

$$\frac{I_{S}}{I_{O}} = 2.71 \times 10^{-13} \tag{7}$$

From equations (4),

$$\frac{I_S}{I_O} = \frac{a_S}{a_O} \frac{A_O}{A_S} \tag{8}$$

and for air at 1 atmosphere, the experimental ratio was found to be

$$\frac{\mathbf{a_S}}{\mathbf{a_O}} = 0.655 \tag{9}$$

Then

$$2.71 \times 10^{-13} = 0.655 \frac{A_0}{A_S} \tag{10}$$

whence

$$\frac{A_0}{A_S} = 4.14 \times 10^{-13} \tag{11}$$

the required instrument constant.

Results

From equation (3) it is noted that the term in brackets is constant for these experiments at a reference pressure of 1 atmosphere. Then,

 $\frac{I_{\mathbf{S}}}{I_{\mathbf{O}}} = \alpha^2 \left[\frac{8\pi^4 N \left(1 + \cos^2 \theta \right)}{r^2 \lambda^4} \right]$ $\frac{I_{\mathbf{S}}}{I_{\mathbf{O}}} = \alpha^2 \ 0.846 \times 10^{35}$ (12)

or

On this basis, the experimental results in terms of α are compared in table I with values of α computed from equation (2) based on optical determinations of refractive index, all at a pressure of 1 atmosphere. The experimental data and values of refractive index used are given in table II.

The linearity of the variation of pressure with the ratio of scattered to incident signals, taken with the excellent agreement found between values of α determined experimentally and those calculated from equation (2) by using values of refractive index determined optically by others, leads to the conclusion that Rayleigh scattering prevails in these experiments.

TABLE I.- EXPERIMENTAL VALUES OF $\, \, lpha \,$ COMPARED WITH COMPUTED VALUES

Gas	α , experimental, $c_{ m m}3$	lpha, computed from optical values of m, cm ³
Air*	1.79×10^{-24}	1.79×10^{-24}
Oxygen, O ₂	1.77	1.67
Nitrogen, N2	1.91	1.82
Helium, He	.217	.221
Argon, A	1.79	1.72
Carbon dioxide, CO2	2.99	3.00

^{*}Standardizing values.

TABLE II.- EXPERIMENTAL DATA AND VALUES
OF REFRACTIVE INDEX

Gas	$a_{\rm S}/a_{\rm O}$, experimental	Index of refraction (optical)
Air	0.655	1.000293
Oxygen, O ₂	.640	1.000272
Nitrogen, N2	.742	1.000297
Helium, He	.0096	1.000036
Argon, A	.655	1.000281
Carbon dioxide, CO2	1.822	1.000490

Other Work

George, et al. (refs. 7 and 8) in experiments similar to these attempt absolute calibration of their apparatus. Their results, expressed in terms of scattering cross section, do not agree with their calculated values by factors ranging between about 1.7 and 2.8 depending upon the test gas. They conclude that this difference is real and consequently, that existing "linear" theory is inadequate. Results of experiments reported herein (see table I) show no differential variability from one gas to another.

Because of the tremendous difficulties attending absolute calibrations of such systems, it was decided in these experiments not to attempt this, but rather, to devise a

relative system and evaluate the system constant by comparison of the experimental values for air under standard conditions with a value computed from well-known and established refractive index of air at optical wavelengths. No significant variation between experiment and calculated values for the various gases was found.

Obviously, one cannot say on the basis of the foregoing that the results of George, et al. are incorrect. It is believed, however, that an "absolute" calibration can be inaccurate by a factor of two or more even when careful attention is given to all facets of the work

Watson, et al. (ref. 9), in a similar experiment, find good agreement between their experiments and values calculated from published indices of refraction for the test gases. They used essentially a relative system and did not attempt absolute calibrations. Their results are reported as ratios between the gases and they did not evaluate their instrument constant.

ADDITIONAL CONSIDERATIONS

Mie Scattering

In general, the hypersonic facilities develop a free stream which is contaminated; that is, it contains other than the natural gases of the atmosphere. If the contaminants are in gaseous form, little difficulty is to be expected in applying the light scattering technique to measurements of gas density. But if the particles are of the order of size equal to or larger than the exploring wavelength, these particles may be expected to give rise to Mie scattering in addition to the normally present Rayleigh scattering.

Although the laboratory experiments performed herein have established the feasibility of determining gas density at selected small volume positions in a gas stream in short time intervals over the range of experimental conditions described, there are additional considerations to be dealt with before a prototype apparatus can be built for application to the facilities.

Probably, the presence of large particles imbedded in the gas stream of hypersonic facilities will generate problems of importance in any application of the technique to simulators. The theory of large particle scattering was first given by Mie (ref. 10). Penndorf (ref. 11) has made a study of Mie scattering for certain particle sizes. Clearly, from Penndorf's work the radiation scattered by large particles is much greater in magnitude than that scattered by gas molecules or atoms; thus, if large particles occur in the free stream, attempts to measure gas density by means of light scattering may well be unsatisfactory unless some way can be found to sort out the kinds of scattering occurring. The variation of Mie scattering with particle size, refractive index of the particles, and

scattering angle approximates a damped sine function and there seems no way of arranging the system geometry to favor one type of scattering over the other.

It may be possible to separate Mie scattering from other types by the use of polarizers and analyzers to take advantage of differences in polarization, and the use of some kind of amplitude sorting on the assumption that the large particles are a small percentage of the total and can be identified by their infrequent occurrence.

Camac, et al. (ref. 12), in their study of laser light scattered from the wake of ballistic pellets, observed that dust in the wake had a pronounced influence on the measurements. They also noted that with an illumination time of the order of 500 microseconds, the individual dust particles could be identified and that in between the large amplitude scattering from these dust particles, the signal level dropped to that caused by Rayleigh scattering from the gas molecules.

Thomson Scattering

Another consideration in this type of experiment involves the presence of free electrons in the gas. Watson, et al. (ref. 9) have calculated that if one atom or molecule in a thousand is ionized, the amplitude of Thomson scattering (scattering of electromagnetic radiation by free electrons) will approximate that of Rayleigh scattering from the remaining unionized particles. This effect has not been examined in these experiments, but such a study should be done to verify the prediction.

Thomson scattering is wavelength independent whereas Rayleigh scattering is inversely proportional to the fourth power of the wavelength. Thus, if several wavelengths can be used, it should be possible to isolate Rayleigh and Thomson scattering from one another.

Differential Drift

Probably, the two most troublesome aspects of the laboratory work were (a) monitoring the laser output, and (b) the differential drift of the photomultiplier tube systems. The first of these can probably be overcome by more elegant monitoring systems, several of which are available. However, both (a) and (b) are related in such a way that if two separate sensors are used, differential drift is likely always to be present. It would seem best to use only one photomultiplier tube both to monitor the laser output and record the scattered light. This could be done if an optical delay line were to be introduced in the monitor train to delay the laser pulse from the monitor and present it to the same phototube used for measuring the scattered light just after the scattered light pulse was finished. When Q-switched lasers are used, the pulse length is of the order of 20×10^{-9} second or less, and the required delay line length then becomes a reasonable

several meters. Such a line could be constructed by using a fiber optics system, since the initial pulse is too strong and needs to be attenuated in any event before entering the photomultiplier tube.

CONCLUSIONS

It appears from the foregoing that proof of feasibility has been established for measuring gas number density by means of laser light scattered by the gas. It is concluded that Rayleigh scattering prevails for these experiments and that the polarizability for each gas is constant over the test range of pressure. Good agreement is found between values of polarizability determined experimentally and those computed from refractive index of the test gases. No differential variability between the polarizability of the gases was found, as had been reported by some other investigators. It is suggested that only one photomultiplier tube should be used to sense both the incident and the scattered light signals; this can be done with an optical delay line in the monitor train.

The technique seems applicable to uncontaminated known gases or gas mixtures over a pressure range of about 50 torr to over 1 atmosphere. Error of pressure measurement with this laboratory apparatus (neglecting helium) was found to average ±5 percent at 600 torr and ±18 percent at 50 torr. Density measurements can be made in about 800 microseconds with a conventional laser and in as short a time as 20 nanoseconds with a Q-switched laser. Volume size for point determinations can be of the order of 0.25 cm³ or less. Further work aimed at perfecting the technique and resolving questions raised in the experiments needs to be done.

Langley Research Center,

National Aeronautics and Space Administration, Langley Station, Hampton, Va., December 1, 1966, 125-24-01-01-23.

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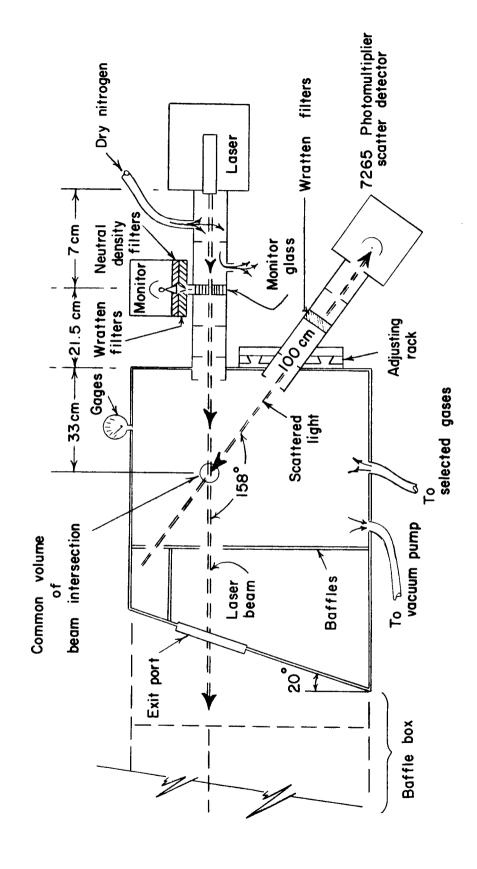


Figure 1.- Sketch of laboratory apparatus for light scattering experiment.

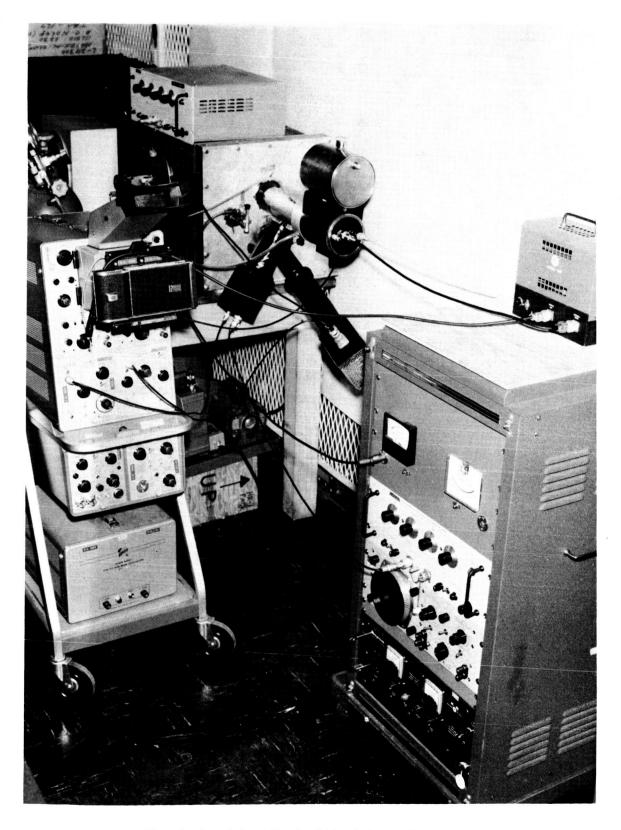


Figure 2.- General view of laboratory light scattering apparatus.

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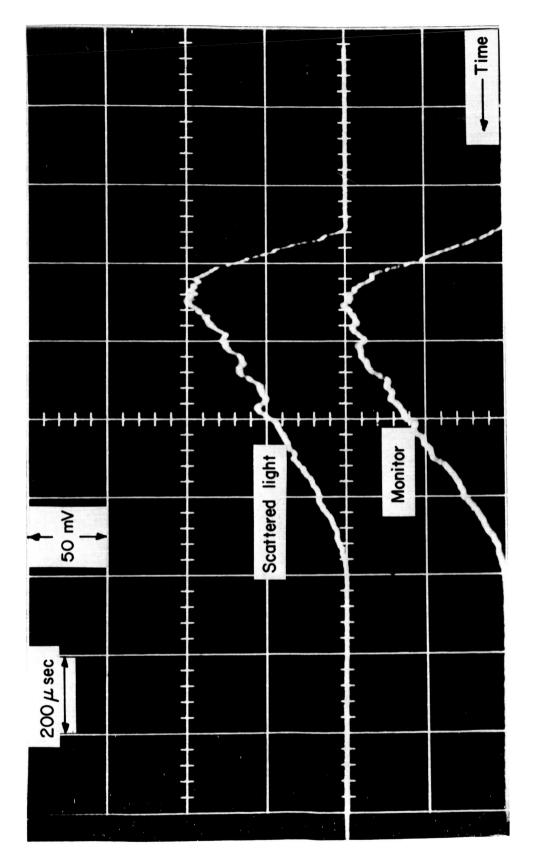
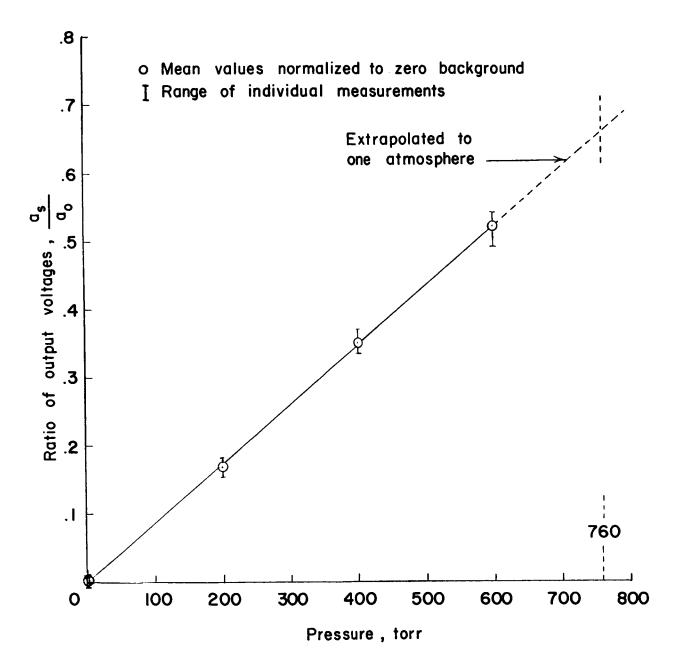
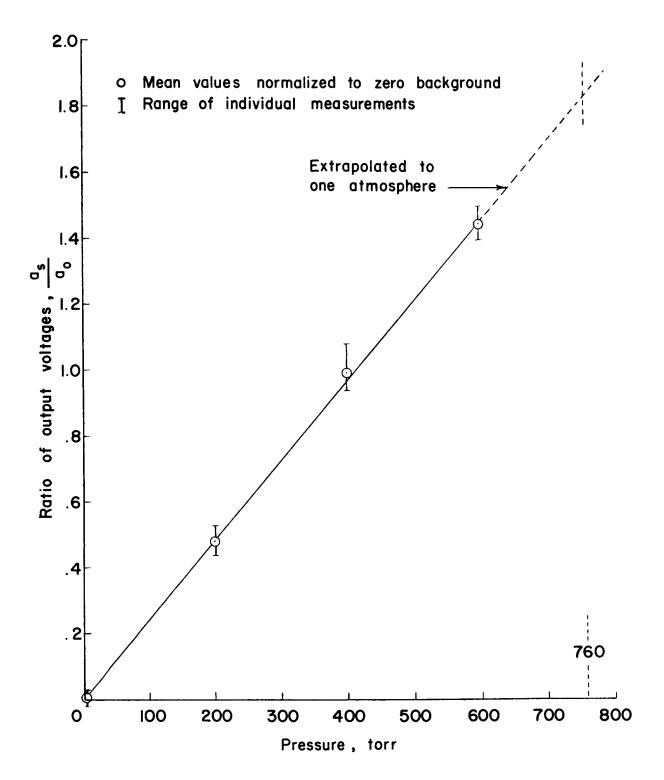


Figure 3.- Laser light scattered from carbon dioxide at 400-mm mercury.



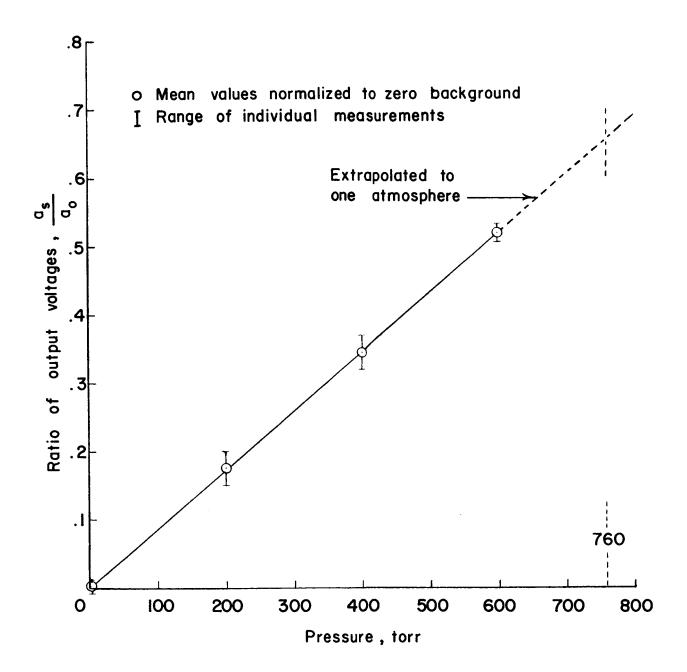
(a) Filtered natural air.

Figure 4.- Ratio of output voltages with pressure.



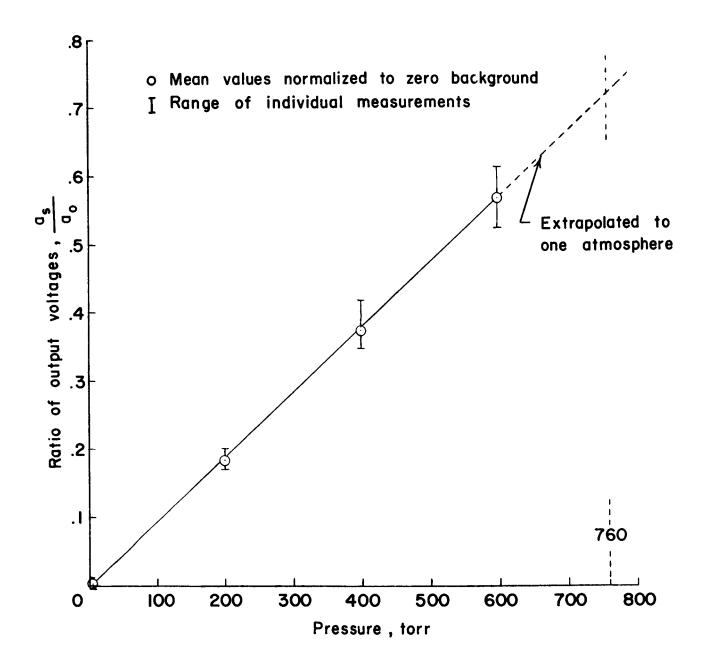
(b) Carbon dioxide.

Figure 4.- Continued.



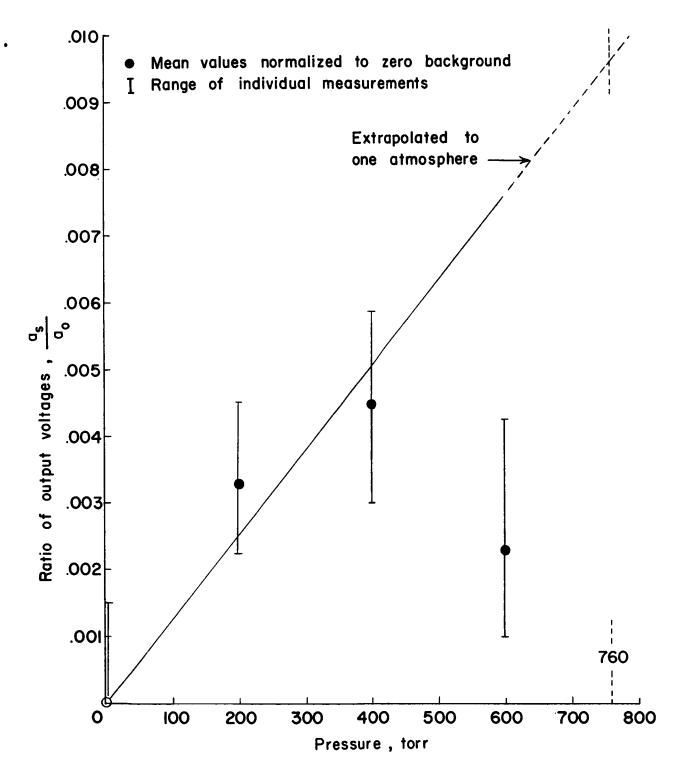
(c) Argon.

Figure 4.- Continued.



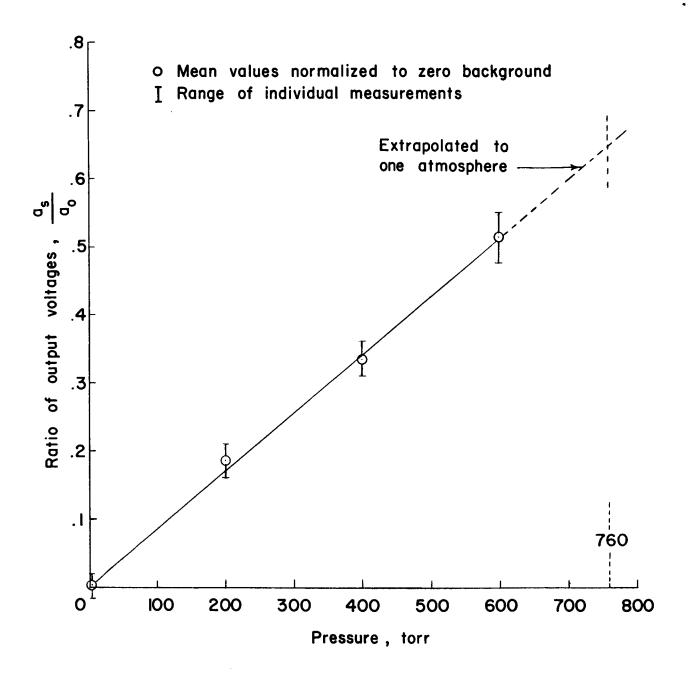
(d) Nitrogen.

Figure 4.- Continued.



(e) Helium.

Figure 4.- Continued.



(f) Oxygen.

Figure 4.- Concluded.

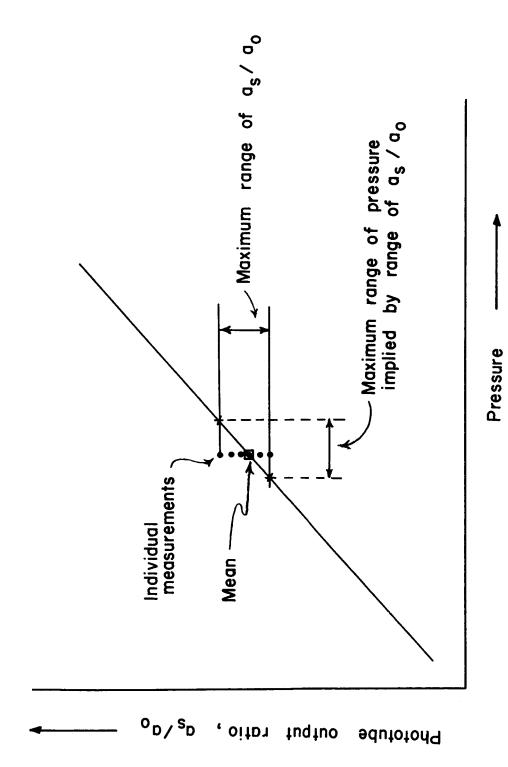


Figure 5.- Relationship between electronic and pressure scatter.

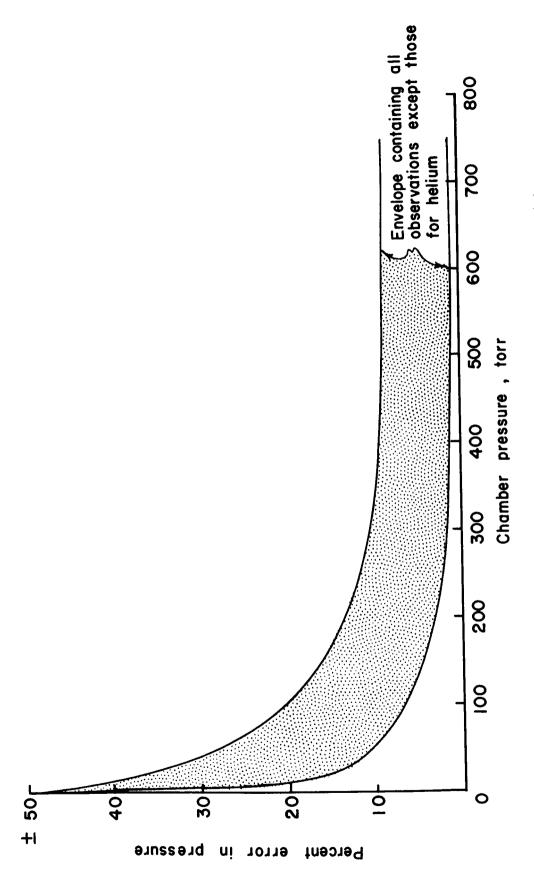


Figure 6.- Variation of error in pressure with pressure deduced from ratio of phototube outputs.

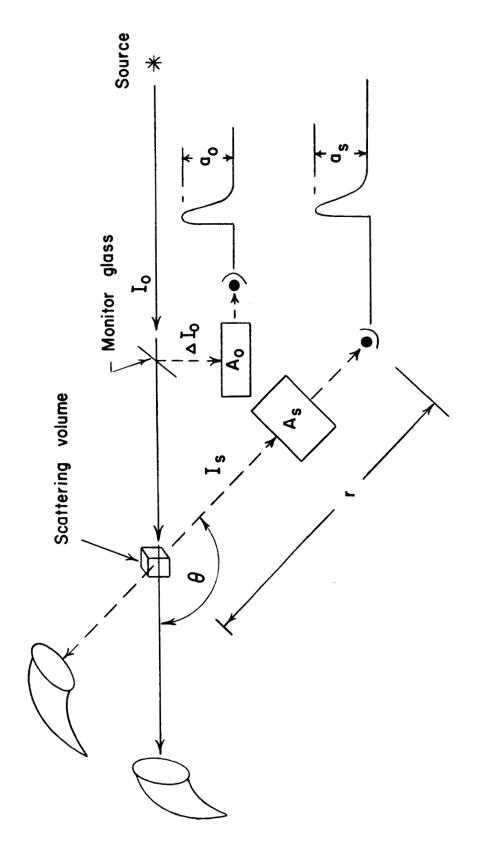


Figure 7.- Schematic diagram of experimental system.

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-NATIONAL AERONAUTICS AND SPACE ACT OF 1958

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